

WHAT IS CLAIMED IS:

1. A two-photon absorbing polymerization method comprising: a first step of irradiating light capable of a two-photon absorption to form a latent image; and a second step of exciting the latent image to cause a polymerization.

2. The two-photon absorbing polymerization method as claimed in claim 1, wherein the excitation in the second step is performed by at least one of a irradiation of light and an application of heat.

3. A two-photon absorbing optical recording method comprising: a first step of forming a latent image of a color-forming material by a two-photon absorption; a second step of irradiating light on said latent image of a color-forming material to cause a polymerization based on a linear absorption of the color-forming material; and thereby forming difference in the refractive index to perform a recording.

4. The two-photon absorbing optical recording method as claimed in claim 3, wherein in said second step, light is irradiated on said latent image of a color-forming material to cause a polymerization while self-sensitizing and self-amplifying based on a linear absorption of the

color-forming material and thereby difference in the refractive index is formed to perform a recording.

5. A two-photon absorbing optical recording material comprising:

1) a two-photon absorbing compound capable of undergoing a two-photon absorption to produce an excited state upon irradiation with light having a wavelength that is longer than the linear absorption band of the compound 1) itself and has a molar absorption coefficient of linear absorption of 10 or less;

2) a dye precursor having an absorption shifted to the longer wavelength side than in the original state by electron or energy transfer from said two-photon absorbing compound 1) in the excited state to become a color-forming material having an absorption in the wavelength region where the molar absorption coefficient of linear absorption in the two-photon absorbing compound 1) is 5,000 or less;

3) a polymerization initiator capable of initiating a polymerization of a polymerizable compound by electron or energy transfer from said two-photon absorbing compound 1) in the excited state;

4) a polymerizable compound; and

5) a binder.

6. The two-photon absorbing optical recording

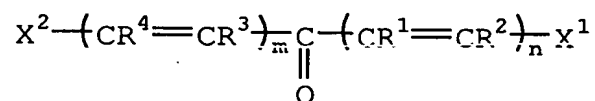
method as claimed in claim 3, wherein the recording is performed by using the two-photon absorbing optical recording material described in claim 5.

7. The two-photon absorbing optical recording method as claimed in claim 6, wherein the wavelength of light for performing the formation of a latent image by two-photon absorption in the first step and the wavelength of light for causing a polymerization by the latent image in the second step are the same.

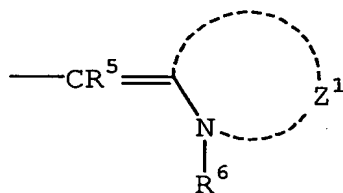
8. The two-photon absorbing optical recording method as claimed in claim 6, wherein the wavelength of light for causing a polymerization by a latent image in the second step is shorter than the wavelength of light for performing the formation of the latent image by two-photon absorption in the first step, and is present in the wavelength region where the molar absorption coefficient of linear absorption in the two-photon absorbing compound is 5,000 or less.

9. The two-photon absorbing polymerization method as claimed in claim 1, wherein as a two-photon absorbing compound, a cyanine dye, a merocyanine dye, an oxonol dye, a phthalocyanine dye or a compound represented by the following formula (1) is used:

Formula (1):



wherein  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  each independently represents a hydrogen atom or a substituent and some of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  may combine with each other to form a ring;  $n$  and  $m$  each independently represents an integer of 0 to 4 and when  $n$  and  $m$  each is 2 or more, multiple  $R^1$ s,  $R^2$ s,  $R^3$ s or  $R^4$ s may be the same or different, provided that  $n$  and  $m$  are not 0 at the same time; and  $X^1$  and  $X^2$  each independently represents an aryl group, a heterocyclic group or a group represented by formula (2):

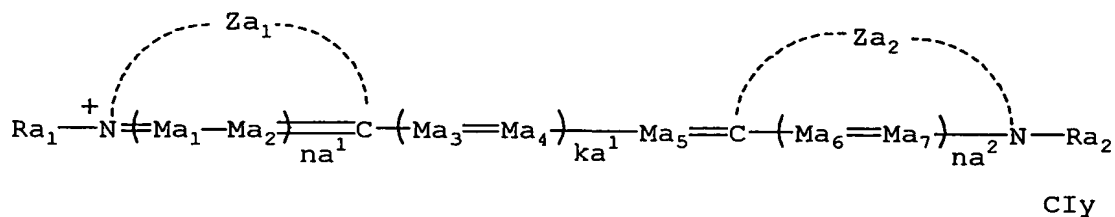


wherein  $R^5$  represents a hydrogen atom or a substituent,  $R^6$  represents a hydrogen atom, an alkyl group, an alkenyl group, an aryl group or a heterocyclic group, and  $Z^1$  represents an atomic group for forming a 5- or 6-membered ring.

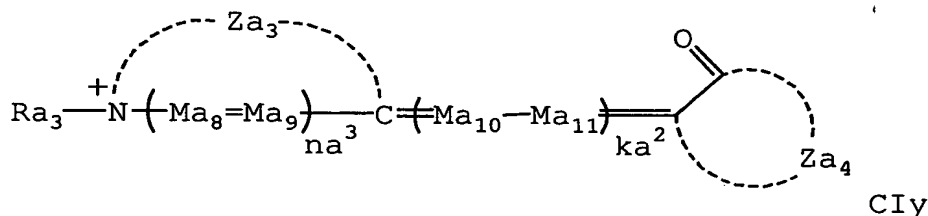
10. The two-photon absorbing polymerization method as claimed in claim 9, wherein the cyanine dye is represented by the following formula (3), the merocyanine

dye is represented by formula (4) and the oxonol dye is represented by formula (5):

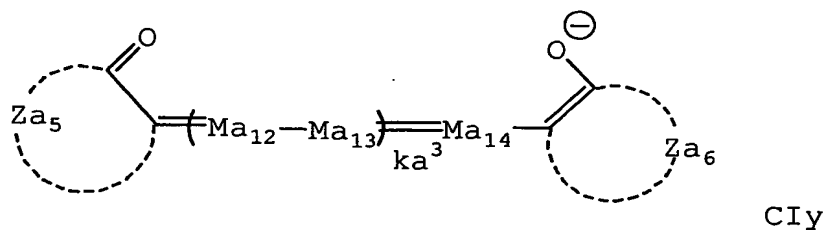
Formula (3):



Formula (4):



Formula (5):

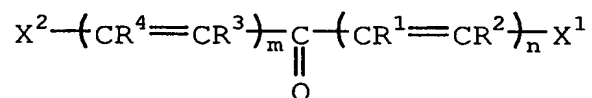


wherein  $\text{Za}_1$ ,  $\text{Za}_2$  and  $\text{Za}_3$  each represents an atomic group for forming a 5- or 6-membered nitrogen-containing heterocyclic ring,  $\text{Za}_4$ ,  $\text{Za}_5$  and  $\text{Za}_6$  each represents an atomic group for forming a 5- or 6-membered ring,  $\text{Ra}_1$ ,  $\text{Ra}_2$  and  $\text{Ra}_3$  each independently represents a hydrogen atom, an alkyl group, an alkenyl group, an aryl group or a heterocyclic group,  $\text{Ma}_1$  to  $\text{Ma}_{14}$  each independently represents a methine group which may have a substituent or may form a ring together with another methine group,  $\text{na}^1$ ,  $\text{na}^2$  and  $\text{na}^3$  each represents

0 or 1,  $ka^1$  and  $ka^3$  each represents an integer of 0 to 3, provided that when  $ka^1$  is 2 or more, multiple  $Ma_{3s}$  or  $Ma_{4s}$  may be the same or different and when  $ka^3$  is 2 or more, multiple  $Ma_{12s}$  or  $Ma_{13s}$  may be the same or different,  $ka^2$  represents an integer of 0 to 8, provided that when  $ka^2$  is 2 or more, multiple  $Ma_{10s}$  or  $Ma_{11s}$  may be the same or different, CI represents an ion for neutralizing the electric charge, and y represents a number necessary for the neutralization of electric charge.

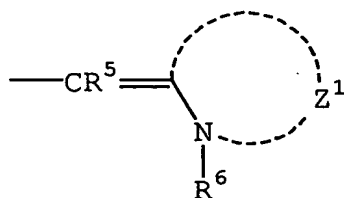
11. The two-photon absorbing optical recording method as claimed in claim 3, wherein as a two-photon absorbing compound, a cyanine dye, a merocyanine dye, an oxonol dye, a phthalocyanine dye or a compound represented by the following formula (1) is used:

Formula (1):



wherein  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  each independently represents a hydrogen atom or a substituent and some of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  may combine with each other to form a ring; n and m each independently represents an integer of 0 to 4 and when n and m each is 2 or more, multiple  $R^1s$ ,  $R^2s$ ,  $R^3s$  or  $R^4s$  may be the same or different, provided that n and m are not 0 at the same time; and  $X^1$  and  $X^2$  each independently represents an aryl group, a heterocyclic group or a group

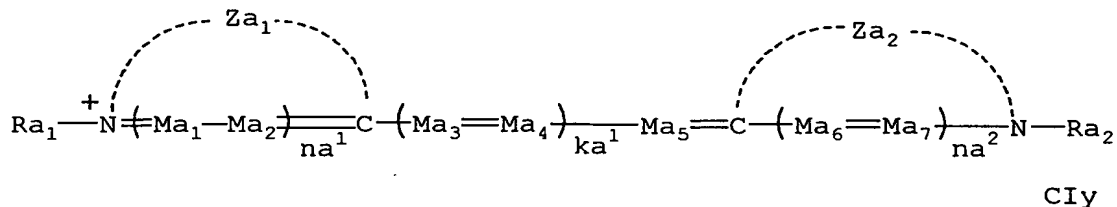
represented by formula (2):



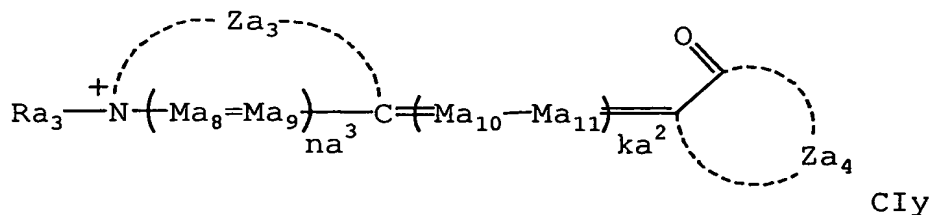
wherein  $R^5$  represents a hydrogen atom or a substituent,  $R^6$  represents a hydrogen atom, an alkyl group, an alkenyl group, an aryl group or a heterocyclic group, and  $Z^1$  represents an atomic group for forming a 5- or 6-membered ring.

12. The two-photon absorbing optical recording method as claimed in claim 11, wherein the cyanine dye is represented by the following formula (3), the merocyanine dye is represented by formula (4) and the oxonol dye is represented by formula (5):

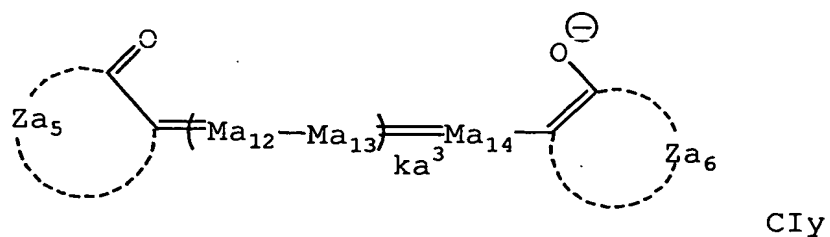
Formula (3):



Formula (4):



Formula (5):



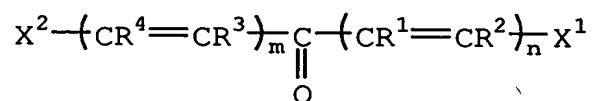
wherein  $Za_1$ ,  $Za_2$  and  $Za_3$  each represents an atomic group for forming a 5- or 6-membered nitrogen-containing heterocyclic ring,  $Za_4$ ,  $Za_5$  and  $Za_6$  each represents an atomic group for forming a 5- or 6-membered ring,  $Ra_1$ ,  $Ra_2$  and  $Ra_3$  each independently represents a hydrogen atom, an alkyl group, an alkenyl group, an aryl group or a heterocyclic group,  $Ma_1$  to  $Ma_{14}$  each independently represents a methine group which may have a substituent or may form a ring together with another methine group,  $na^1$ ,  $na^2$  and  $na^3$  each represents 0 or 1,  $ka^1$  and  $ka^3$  each represents an integer of 0 to 3, provided that when  $ka^1$  is 2 or more, multiple  $Ma_3$ s or  $Ma_4$ s may be the same or different and when  $ka^3$  is 2 or more, multiple  $Ma_{12}$ s or  $Ma_{13}$ s may be the same or different,  $ka^2$  represents an integer of 0 to 8, provided that when  $ka^2$  is 2 or more, multiple  $Ma_{10}$ s or  $Ma_{11}$ s may be the same or different, CI represents an ion for neutralizing the electric charge, and  $y$  represents a number necessary for the neutralization of electric charge.

13. The two-photon absorbing optical recording material as claimed in claim 5, wherein said two-photon

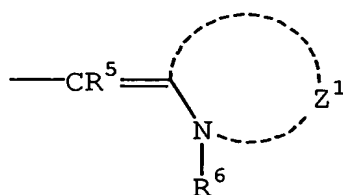


absorbing compound is a cyanine dye, a merocyanine dye, an oxonol dye, a phthalocyanine dye or a compound represented by the following formula (1):

Formula (1):



wherein  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  each independently represents a hydrogen atom or a substituent and some of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  may combine with each other to form a ring;  $n$  and  $m$  each independently represents an integer of 0 to 4 and when  $n$  and  $m$  each is 2 or more, multiple  $R^1$ 's,  $R^2$ 's,  $R^3$ 's or  $R^4$ 's may be the same or different, provided that  $n$  and  $m$  are not 0 at the same time; and  $X^1$  and  $X^2$  each independently represents an aryl group, a heterocyclic group or a group represented by formula (2):

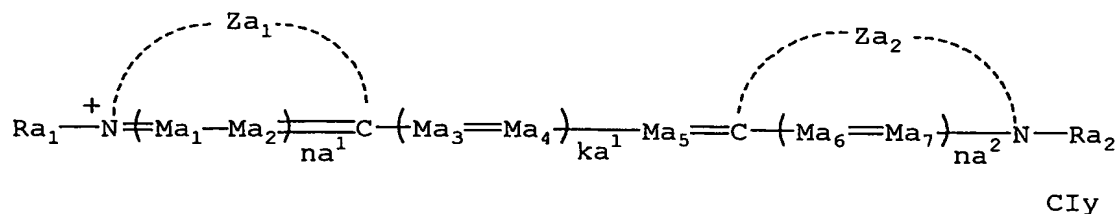


wherein  $R^5$  represents a hydrogen atom or a substituent,  $R^6$  represents a hydrogen atom, an alkyl group, an alkenyl group, an aryl group or a heterocyclic group, and  $Z^1$  represents an atomic group for forming a 5- or 6-membered ring.

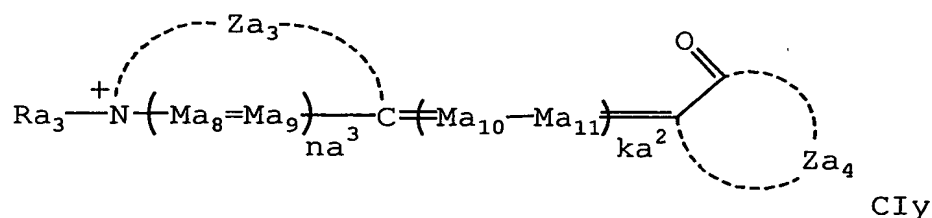
14. The two-photon absorbing optical recording

material as claimed in claim 13, wherein the cyanine dye is represented by the following formula (3), the merocyanine dye is represented by formula (4) and the oxonol dye is represented by formula (5):

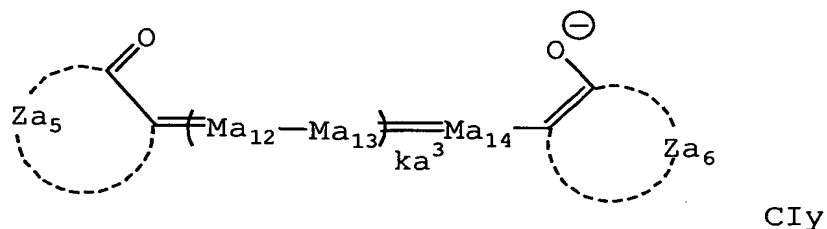
Formula (3):



Formula (4):



Formula (5):



wherein  $\text{Za}_1$ ,  $\text{Za}_2$  and  $\text{Za}_3$  each represents an atomic group for forming a 5- or 6-membered nitrogen-containing heterocyclic ring,  $\text{Za}_4$ ,  $\text{Za}_5$  and  $\text{Za}_6$  each represents an atomic group for forming a 5- or 6-membered ring,  $\text{Ra}_1$ ,  $\text{Ra}_2$  and  $\text{Ra}_3$  each independently represents a hydrogen atom, an alkyl group, an alkenyl group, an aryl group or a heterocyclic group,  $\text{Ma}_1$  to  $\text{Ma}_{14}$  each independently represents a methine group

which may have a substituent or may form a ring together with another methine group,  $na^1$ ,  $na^2$  and  $na^3$  each represents 0 or 1,  $ka^1$  and  $ka^3$  each represents an integer of 0 to 3, provided that when  $ka^1$  is 2 or more, multiple  $Ma_3s$  or  $Ma_4s$  may be the same or different and when  $ka^3$  is 2 or more, multiple  $Ma_{12}s$  or  $Ma_{13}s$  may be the same or different,  $ka^2$  represents an integer of 0 to 8, provided that when  $ka^2$  is 2 or more, multiple  $Ma_{10}s$  or  $Ma_{11}s$  may be the same or different, CI represents an ion for neutralizing the electric charge, and y represents a number necessary for the neutralization of electric charge.

15. A photon absorbing optical recording and reproducing method comprising: performing a recording by the first and second steps described in claim 3; then performing a reproduction by irradiating light on said recorded area and detecting the difference in reflectance attributable to the difference in refractive index.

16. The two-photon absorbing optical recording method as claimed in claim 6, wherein the polymerizable compound and the binder have a difference in the refractive index, and the compositional ratio of the polymerizable compound and a polymerization reaction product thereof to the binder becomes non-uniform between the recorded area and the unrecorded area to enable a two-photon absorbing

optical recording by utilizing a modulation of refractive index.

17. The two-photon absorbing optical recording material as claimed in claim 5, wherein the polymerizable compound contains at least one selected from the group consisting of an aryl group, an aromatic heterocyclic group, a chlorine atom, a bromine atom, an iodine atom and a sulfur atom, and the binder has a refractive index lower than that of the polymerizable compound.

18. The two-photon absorbing optical recording material as claimed in claim 5, wherein the dye precursor is an acid-color forming dye precursor or a base-color forming dye precursor.

19. A two-photon absorbing polymerizable composition comprising a two-photon absorbing compound, a polymerization initiator, a polymerizable compound and a binder, in which the two-photon absorbing polymerizable composition is capable of generating a three-dimensional modulation of refractive index as a result of photopolymerization caused by non-resonant two-photon absorption.

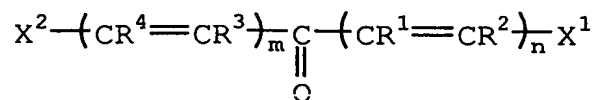
20. A photon absorbing optical recording and reproducing method comprising: performing a recording by

using the two-photon absorbing polymerizable composition described in claim 19; then performing a reproduction by irradiating light on said recorded area and detecting the difference in reflectance attributable to the difference in refractive index.

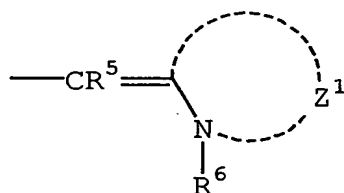
21. The two-photon absorbing polymerizable composition as claimed in claim 19, wherein the polymerizable compound and the binder have a difference in the refractive index, and the compositional ratio of the polymerizable compound and a polymerization reaction product thereof to the binder becomes non-uniform between the recorded area and the unrecorded area to enable a two-photon absorbing optical recording by utilizing a modulation of refractive index.

22. The two-photon absorbing polymerizable composition as claimed in claim 19, wherein said two-photon absorbing compound is a methine dye or a phthalocyanine dye.

23. The two-photon absorbing polymerizable composition as claimed in claim 19, wherein said two-photon absorbing compound is a cyanine dye, a merocyanine dye, an oxonol dye, a phthalocyanine dye or a compound represented by the following formula (1):



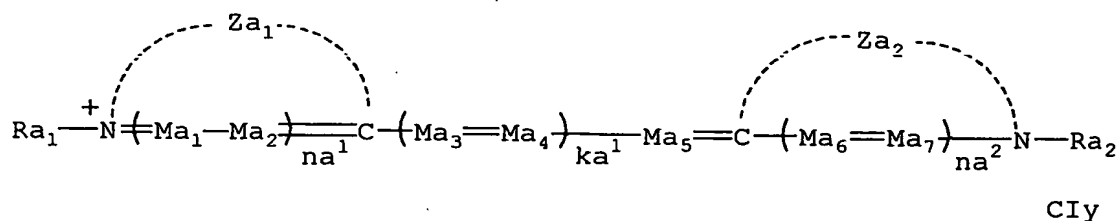
wherein  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  each independently represents a hydrogen atom or a substituent and some of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  may combine with each other to form a ring;  $n$  and  $m$  each independently represents an integer of 0 to 4 and when  $n$  and  $m$  each is 2 or more, multiple  $R^1$ s,  $R^2$ s,  $R^3$ s or  $R^4$ s may be the same or different, provided that  $n$  and  $m$  are not 0 at the same time; and  $X^1$  and  $X^2$  each independently represents an aryl group, a heterocyclic group or a group represented by formula (2):



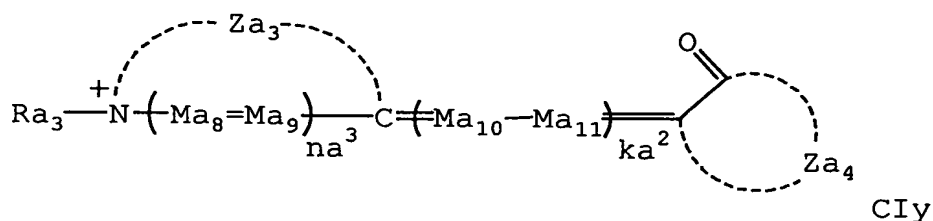
wherein  $R^5$  represents a hydrogen atom or a substituent,  $R^6$  represents a hydrogen atom, an alkyl group, an alkenyl group, an aryl group or a heterocyclic group, and  $Z^1$  represents an atomic group for forming a 5- or 6-membered ring.

24. The two-photon absorbing polymerizable composition as claimed in claim 23, wherein the cyanine dye is represented by the following formula (3), the merocyanine dye is represented by formula (4) and the oxonol dye is represented by formula (5):

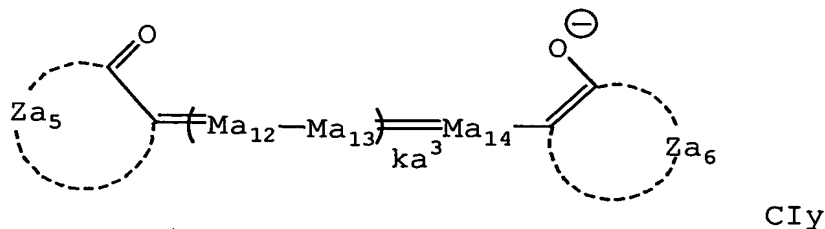
Formula (3):



Formula (4):



Formula (5):



wherein  $\text{Za}_1$ ,  $\text{Za}_2$  and  $\text{Za}_3$  each represents an atomic group for forming a 5- or 6-membered nitrogen-containing heterocyclic ring,  $\text{Za}_4$ ,  $\text{Za}_5$  and  $\text{Za}_6$  each represents an atomic group for forming a 5- or 6-membered ring,  $\text{Ra}_1$ ,  $\text{Ra}_2$  and  $\text{Ra}_3$  each independently represents a hydrogen atom, an alkyl group, an alkenyl group, an aryl group or a heterocyclic group,  $\text{Ma}_1$  to  $\text{Ma}_{14}$  each independently represents a methine group which may have a substituent or may form a ring together with another methine group,  $\text{na}^1$ ,  $\text{na}^2$  and  $\text{na}^3$  each represents 0 or 1,  $\text{ka}^1$  and  $\text{ka}^3$  each represents an integer of 0 to 3, provided that when  $\text{ka}^1$  is 2 or more, multiple  $\text{Ma}_3$ s or  $\text{Ma}_4$ s

may be the same or different and when  $ka^3$  is 2 or more, multiple  $Ma_{12}s$  or  $Ma_{13}s$  may be the same or different,  $ka^2$  represents an integer of 0 to 8, provided that when  $ka^2$  is 2 or more, multiple  $Ma_{10}s$  or  $Ma_{11}s$  may be the same or different, CI represents an ion for neutralizing the electric charge, and y represents a number necessary for the neutralization of electric charge.

25. A method for three-dimensionally modulating a refractive index, comprising: irradiating the two-photon absorbing polymerizable composition described in claim 19 with laser light at a wavelength being longer than the linear absorption band of the two-photon absorbing compound and having no linear absorption to induce a two-photon absorption; and causing photopolymerization by utilizing the two-photon absorption induced.

26. A three-dimensional optical recording method comprising:

irradiating the two-photon absorbing polymerizable composition described in claim 19 with laser light at a wavelength being longer than the linear absorption band of the two-photon absorbing compound and having no linear absorption to induce a two-photon absorption;

causing photopolymerization by utilizing the two-photon absorption induced to cause a non-uniformity of the



compositional ratio of the polymerizable compound and a polymerization reaction product thereof to the binder between the laser-focused area and unfocused area; and

performing a recording by using a three-dimensional modulation of refractive index caused by the non-uniformity of the compositional ratio.